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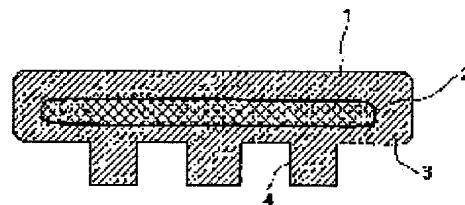
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(54) SEPARATOR FOR FUEL CELL AND SOLID POLYMER FUEL CELL

(57)Abstract:

PROBLEM TO BE SOLVED: To prevent strength and gas impermeability from being lowered even thinning a separator by bonding a conductive core part formed of a metallic material or metallic composite material, to a conductive skin part with conductive adhesive layer.

SOLUTION: A conductive core part 1 is formed of metallic material represented by titanium, aluminum and stainless steel or metallic composite material formed by coating the metallic material with noble metal or carbon material. In order to heighten adhesion to a conductive skin part 3, blasting, electric discharge or a surface treatment by a mechanical polishing method such as lapping or polishing may be applied to the conductive core part 1. A conductive adhesive layer 2 for covering the conductive core part 1 is formed using a conductive adhesive. Additives such as a dispersant, a thickener, a stabilizer and a defoaming agent may be added to the conductive adhesive, if necessary.



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CLAIMS

[Claim(s)]

[Claim 1] The separator for fuel cells characterized by having consisted of the conductive core section by the metallic material or metal matrix composite, a conductive glue line which covers this conductive core section, and the conductive skin section formed on this conductive glue line, and the conductive core section and the conductive skin section having pasted up by the conductive glue line.

[Claim 2] A conductive glue line is a separator for fuel cells according to claim 1 currently formed by the carbon content electroconductive glue which makes carbon powder a conductive filler and uses resin as a binder.

[Claim 3] The conductive skin section is a separator for fuel cells according to claim 1 currently formed with the carbon content composite material which makes carbon powder a conductive filler and uses resin as a binder.

[Claim 4] The polymer electrolyte fuel cell which comes to use the separator for fuel cells according to claim 1 to 3.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the separator for fuel cells without ** Li and the reinforcement and gas impermeability falling, even if it sheet-metal-izes in more detail about the separator for fuel cells.

[0002]

[Description of the Prior Art] The separator for fuel cells drains the water generated all over said slot in process of the generation of electrical energy, and is bearing the role of securing the slot concerned as a circulation way of the reactant gas which flows into a fuel cell cel while it transmits to the exterior the electrical and electric equipment which comes to form the slot where plurality is generally parallel to monotonous both sides or monotonous one side, and was generated with the gas diffusion electrode in a fuel cell cel.

[0003] Although sheet-metal-izing also about the above separators for fuel cells came to be called for with lightweight-izing and thin-shape-izing of a fuel cell in recent years, when only sheet-metal-ized, reinforcement fell, and the conventional separator for fuel cells had the problem that gas permeability will become high.

[0004] In order to solve the above-mentioned problem, mold shaping of the mixture of carbon powder and phenol resin is carried out, and the separator for fuel cells of the structure which contains a metal mesh or the piece of a metal in a core (refer to JP,59-149657,A), and the separator for fuel cells (refer to U.S. Pat. No. 5,527,363) obtained by hammering out a gas passageway at the same time it inserts a metal sheet between compressible conductive liner sheets and is stuck by pressure, putting a pressure are proposed.

[0005]

[Problem(s) to be Solved by the Invention] However, there is the same problem as the impact at the time of the assembly of a fuel cell and the conventional thing that a metal mesh etc. will tend to exfoliate within mold mold goods, and conductivity and gas permeability will become high with external pressure, such as bolting, in the separator for fuel cells which contains a metal mesh etc. with the above-mentioned carbon powder, phenol resin, and mold mold goods in a core.

[0006] moreover, to the separator for fuel cells obtained by inserting a metal sheet between compressible conductive liner sheets, and being stuck by pressure, putting a pressure Since the conductive liner sheet and the metal sheet are only stuck by pressure, bond strength is inadequate. This sake, Since the adhesion of about [that there is a possibility of producing the problem that they will exfoliate on the occasion of use in hot water], a conductive liner sheet, and a metal sheet is bad There is a difficulty that the contact resistance between a conductive liner sheet and a metal sheet becomes high, the resistance as a separator for fuel cells is high, and especially aging in the inside of hot water becomes intense.

[0007] This invention was made for the purpose of offering the separator for fuel cells with which the reinforcement or gas impermeability do not fall even if it cancels and sheet-metal-izes the difficulty of the above conventional techniques.

[0008]

[Means for Solving the Problem] The configuration of the separator for fuel cells which this invention adopted in order to attain the above-mentioned purpose consists of the conductive core section by the metallic material or metal matrix composite, a conductive glue line which covers this conductive core section, and the conductive skin section formed on this conductive glue line, and is characterized by the conductive core section and the conductive skin section having pasted up by the conductive glue line.

[0009]

[The mode of implementation of invention] This invention is explained below at a detail.

[0010] The separator for fuel cells of this invention consists of the conductive core section 1 by the metallic material or metal matrix composite, a conductive glue line 2 which covers this conductive core section 1, and the conductive skin section 3 formed on this conductive glue line 2, as shown in drawing 1 . In addition, 4 is the circulation way of the reactant gas which flows into a fuel cell cel.

[0011] The separator for fuel cells of this invention is not limited to the configuration shown in drawing 1 , for example, may form the circulation way 4 of reactant gas also in the upper part.

[0012] The above-mentioned conductive core section 1 is formed of the metallic material represented by titanium, aluminum, stainless steel, etc. and the metal matrix composite which coated these metallic materials with noble metals, a carbon ingredient, etc.

[0013] What has 5 micrometers in thickness as shown in drawing 1 , the about 3mm sheet-like object, or the configuration corresponding to the separator for fuel cells as a configuration of the above-mentioned conductive core section 1, for example, and the thing which has the configuration corresponding to the separator for fuel cells in the part can be mentioned.

[0014] Moreover, in order to raise an adhesive property with the conductive skin section 3 mentioned later, as opposed to the conductive core section 1, surface treatment by the mechanical polish approaches, such as blasting processing, electrodischarge treatment, wrapping, and polishing, may be performed.

[0015] In drawing 1 , the conductive glue line 2 which covers the conductive core section 1 is formed using the adhesives which have conductivity, and can mention the carbon content electroconductive glue which makes carbon powder a conductive filler and uses resin as a binder as adhesives which have this conductivity, for example.

[0016] Although there will be especially no limit if the carbon powder as a conductive filler in the adhesives which have the above-mentioned conductivity can be used for the purpose which gives conductivity, one kind or two kinds or more of mixture chosen from natural graphites, such as Lynn flaky graphite and a soil massive graphite, an artificial graphite, expanded graphite, acetylene black, carbon black, and KETCHIEN black can be mentioned, for example.

[0017] moreover, although there will be especially no limit as resin as a binder in the adhesives which have the above-mentioned conductivity if it can be use for the purpose which strengthen junction in the skin section and the core section while raise conductivity, for example, one kind or two kinds or more of mixture chose from thermosetting resin, thermoplastics, and rubber can be mention, and these may be liquefied or may be emulsion-like things.

[0018] As the above-mentioned thermosetting resin, one kind or two kinds or more of mixture chosen from phenol resin, poly carbodiimide resin, furfuryl alcohol resin, an epoxy resin, a cellulose, a urea-resin, melamine resin, an unsaturated polyester resin, silicone resin, diallyl phthalate resin, bismaleimide triazine resin, the poly amino bismaleimide resin, and aromatic polyimide resin can be mentioned, for example.

[0019] As the above-mentioned thermoplastics, for example Moreover, polyethylene, polystyrene,

Polypropylene, a polymethyl methacrylate, polyethylene terephthalate, Polybutylene terephthalate, polyether sulfone, a polycarbonate, Poly OKISA methylene, a polyamide, polyimide, polyamidoimide, Polyvinyl alcohol, polyvinyl chloride, the poly FENIRUSARU phon, A polyether ether ketone, polysulfone, a polyether ketone, Polyarylate, polyether imide, the poly methyl pentene, fluororesin, Polyoxybenzoyl ester resin, liquid crystal polyester resin, aromatic polyester, One kind or two kinds or more of mixture chosen from polyacetal, the poly allyl compound sulfone, the poly benzimidazole, polyether nitril, the poly thioether sulfone, and polyphenylene ether can be mentioned.

[0020] Furthermore, as rubber, one kind or two kinds or more of mixture chosen from a fluororubber, silicone rubber, isobutylene isoprene rubber, chloroprene rubber, 2 tolyl rubber, 2 tolyl chloroprene rubber, chlorinated butyl rubber, epichlorohydrin rubber, epichlorohydrin-ethyleneoxide rubber, an epichlorohydrin-ethyleneoxide-acrylic glycol ether three-dimension copolymer, polyurethane rubber, acrylic rubber, ethylene-propylene rubber, a styrene rubber, butadiene rubber, and natural rubber can be mentioned, for example.

[0021] In addition, in the adhesives which have the above-mentioned conductivity, additives, such as a dispersant, a thickener, a stabilizer, and defoaming material, may be added if needed.

[0022] As a quantitative ratio of the carbon powder and resin in the adhesives which have the above-mentioned conductivity, range called resin 10 – the 150 weight sections can be illustrated to the carbon powder 100 weight section. In addition, when the problem that sufficient glue line cannot be obtained when resin is less than 10 weight sections arises and it exceeds the 150 weight sections, the problem that sufficient conductivity cannot be acquired arises and neither is desirable.

[0023] As an approach of applying the electroconductive glue obtained in this way to the conductive core section 1 that what is necessary is just to only mix the above-mentioned carbon powder and resin, and covering this conductive core section 1 with the conductive glue line 2 About the electroconductive glue which could mention the approach that casting, DIP coating, a spray, brush coating, screen-stencil, etc. were conventionally well-known, and was applied in this way In the condition [having applied], you may introduce to a junction process with the conductive skin section mentioned later, or it may put in into a dryer, and desiccation and hardening of may be done. In addition, as a drying temperature in this desiccation process, the range of 30 degrees C – 200 degrees C can be illustrated.

[0024] In addition, when the range of 0.1 micrometers – 200 micrometers can be illustrated as thickness of the conductive glue line 2 and it exceeds this range, sufficient bond strength cannot be obtained and it is not desirable.

[0025] The conductive skin section 3 of this invention aims at reducing contact resistance with the contacted bodies (a membrane electrode, carbon paper, etc.), and can mention the carbon content composite material which constitutes the conductive skin section 3 and which makes carbon powder a conductive filler as a material for reducing contact resistance, and uses resin as a binder.

[0026] Although there will be especially no limit if the carbon powder as a conductive filler in the material for reducing the above-mentioned contact resistance can be used for the purpose which gives conductivity, one kind or two kinds or more of mixture chosen from natural graphites, such as Lynn flaky graphite and a soil massive graphite, an artificial graphite, expanded graphite, acetylene black, carbon black, and KETCHIEN black can be mentioned like the case of the adhesives which have the above-mentioned conductivity.

[0027] moreover, as resin as a binder in the material for reducing the above-mentioned contact resistance Although there will be especially no limit if the reinforcement of the skin section can be used for the improving purpose while raising conductivity One kind or two kinds or more of mixture chosen from thermosetting resin, thermoplastics, and rubber can be mentioned like the case of the adhesives which have the above-mentioned conductivity, and these may be liquefied or may be emulsion-like things.

[0028] As the above-mentioned thermosetting resin, one kind or two kinds or more of mixture chosen from phenol resin, poly carbodiimide resin, furfuryl alcohol resin, an epoxy resin, a cellulose, a urea-resin, melamine resin, an unsaturated polyester resin, silicone resin, diallyl phthalate resin, bismaleimide triazine resin, the poly amino bismaleimide resin, and aromatic polyimide resin can be mentioned, for example.

[0029] As the above-mentioned thermoplastics, for example Moreover, polyethylene, polystyrene, Polypropylene, a polymethyl methacrylate, polyethylene terephthalate, Polybutylene terephthalate, polyether sulfone, a polycarbonate, Poly OKISA methylene, a polyamide, polyimide, polyamidoimide, Polyvinyl alcohol, polyvinyl chloride, the poly FENIRUSARU phon, A polyether ether ketone, polysulfone, a polyether ketone, Polyarylate, polyether imide, the poly methyl pentene, a fluororesin, Polyoxybenzoyl ester resin, liquid crystal polyester resin, aromatic polyester, One kind or two kinds or more of mixture chosen from polyacetal, the poly allyl compound sulfone, the poly benzimidazole, polyether nitril, the poly thioether sulfone, and polyphenylene ether can be mentioned.

[0030] Furthermore, as rubber, one kind or two kinds or more of mixture chosen from a fluororubber, silicone rubber, isobutylene isoprene rubber, chloroprene rubber, nitrile rubber, nitril chloroprene rubber, chlorinated butyl rubber, epichlorohydrin rubber, epichlorohydrin-ethyleneoxide rubber, an epichlorohydrin-ethyleneoxide-

acrylic glyceryl ether three-dimension copolymer, polyurethane rubber, acrylic rubber, ethylene-propylene rubber, a styrene rubber, butadiene rubber, and natural rubber can be mentioned, for example.

[0031] As a quantitative ratio of the carbon powder and resin in the material for reducing the contact resistance which constitutes the conductive skin section 3 of this invention, range called resin 3 – 35 weight sections can be mentioned to the carbon powder 100 weight section. In addition, in the conductive skin section's 3 being unable to maintain sufficient reinforcement when resin is under 3 weight sections, for example, there being a possibility of saying that the conductive skin section 3 will collapse with the generation water at the time of being a fuel cell generation of electrical energy and exceeding 35 weight sections, there is a problem that the conductivity required of the conductive skin section 3 is not securable.

[0032] In addition, the conductive glue line 2 and the conductive skin section 3 can also be made to unify by making into the same thing the material for reducing the adhesives and contact resistance which have the above-mentioned conductivity.

[0033] On the other hand, the fuel cell separator of this invention can be manufactured by at least three kinds of approaches according to the mode which forms the conductive skin section 3.

[0034] The 1st manufacture approach is the approach of producing the conductive skin section 3 beforehand and joining to the conductive core section 1 with electroconductive glue, as shown in drawing 2 . Namely, the 2nd manufacture approach As are shown in drawing 3 , and the conductive core section-skin section zygote which electroconductive glue is applied [zygote] and makes it have rivaled is thrown in in metal mold, it is the approach of fabricating in a separator configuration and the 3rd manufacture approach is shown in drawing 4 It is the approach of throwing in and fabricating the raw material of the conductive skin section on both sides or one side to the conductive core section 1 which has applied electroconductive glue.

[0035] Although the separator for fuel cells is obtained in this invention by pasting up the conductive core section 1 and the conductive skin section 3 by the conductive glue line 2 as above-mentioned, also at this process, a well-known approach can be used conventionally, for example, approaches, such as mold shaping, injection molding, extrusion molding, roll forming, and hydrostatic molding, can be adopted.

[0036] An example explains this invention further below at a detail.

[0037] The example 1 – 5 Lynn flaky graphite (3 micrometers of mean diameters) 50 weight section, the acetylene black (40nm of mean diameters) 50 weight section, and the phenol resin 32 weight section were mixed with the ball mill, and electroconductive glue was produced. Moreover, said electroconductive glue was applied by the spray all over this, electroconductive glue was dried, using a stainless steel plate (plate without 0.1mm in SUS316 and thickness and a slot) as the core section, and the electroconductive glue layer with a thickness of 30 micrometers was formed.

[0038] As shown in drawing 2 , the skin section raw material mixture shown in Table 1 was thrown into the separator molding die which the rib attached, it left the conductive skin section which fabricated and fabricated only the two conductive skin sections beforehand in metal mold, and the near punch or the female mold to which the core section is joined was removed. It equipped with the conductive core section which covered said conductive glue line between the metal mold which left the two fabricated conductive skin sections, shaping was performed for 5 minutes by 152 degrees C and 120kg/cm², and the separator with a thickness of 2mm which has a conductive core layer in a core was manufactured.

[0039] The specific resistance and gas permeability of the separator for fuel cells which were produced were measured, it equipped with the separator for fuel cells further produced to the repeat compression tester currently used by JIS K6400 (flexible-urethane-foam test method), and the condition of the separator after giving 20 repeat compression was observed. Moreover, this fuel cell separator was included in the polymer electrolyte fuel cell, it bound tight by the pressure of 100kg/cm², the fuel cell was generated in this condition for 500 hours, and the voltage drop 500 hours after [when setting an early electrical potential difference to 100] was measured. Moreover, after generating electricity for 500 hours, this fuel cell was disassembled, and the situation of a separator was observed. These trials and an observation result are shown in Table 1.

[0040] In six to example 10 examples 1–5, except having used the aluminum plate (plate without the thickness of 0.1mm and a slot) as a charge of a core member, the fuel cell separator using the skin section raw material mixture shown in Table 1 at the skin section was produced similarly, and it was similarly estimated as examples 1–5. The result is shown in Table 1.

[0041]

[Table 1]

[0044] In example 23 example 4, the conductive skin section which fabricated shaping of a separator to plate-like according to drawing 3 was made to rival the core section by the conductive glue line, the junction precursor was produced, and the separator shaping metal mold which the rib attached next was equipped with this junction precursor, and except having joined the conductive skin section and the core section, the separator for fuel cells was produced and it was similarly estimated as examples 1-5. The result is shown in Table 3.

[0045] In example 24 example 4, one half was supplied to the female mold of the separator shaping metal mold with which the rib attached shaping of a separator according to drawing 4 among the raw materials of the conductive skin section, and the core section which subsequently applied electroconductive glue was supplied, and except having thrown in and fabricated the remaining raw materials of the conductive skin section on it further, the separator for fuel cells was produced and it was similarly estimated as examples 1-5. The result is shown in Table 3.

[0046] Mold shaping was performed using the presentation of the skin section of example of comparison 1 example 2, and the separator for fuel cells with a thickness of 1.0mm was manufactured. Like the example 1, after incorporating into the fuel cell and binding tight, it dissolved, and it observed about the appearance condition of the specific resistance of the separator for fuel cells in that case, gas permeability, an electrical-potential-difference value, and the separator for fuel cells. A result is shown in Table 3.

[0047] Injection molding of the liquid crystal polymer was carried out to both sides of an example of comparison 2 aluminum plate, the separator base material was formed, gold was vapor-deposited on it, and the separator for fuel cells with a thickness of 1.0mm was manufactured. Like the example 1, after incorporating into the fuel cell and binding tight, it dissolved, and it observed about the appearance condition of the specific resistance of the separator for fuel cells in that case, gas permeability, an electrical-potential-difference value, and the separator for fuel cells. A result is shown in Table 3.

[0048] In example of comparison 3 example 1, an aluminum mesh was instead put into the core part, mold shaping was performed, and the separator for fuel cells with a thickness of 1.0mm was manufactured. Like the example 1, after incorporating into the fuel cell and binding tight, it dissolved, and it observed about the appearance condition of the specific resistance of the separator for fuel cells in that case, gas permeability, an electrical-potential-difference value, and the separator for fuel cells. A result is shown in Table 3.

[0049] The expanded graphite sheet was put between both sides of an example of comparison 4 aluminum sheet (1.0mm in thickness), mold shaping was performed, and the separator for fuel cells with a thickness of 1.0mm was manufactured. Like the example 1, after incorporating into the polymer electrolyte fuel cell and binding tight, it dissolved, and it observed about the appearance condition of the specific resistance of the separator for fuel cells in that case, gas permeability, an electrical-potential-difference value, and the separator for fuel cells. A result is shown in Table 3.

[0050]

[Table 3]

		実施例23	実施例24	比較例1	比較例2	比較例3	比較例4
導電性接着剤	天然黒鉛 (平均粒径20 μ m)						
	アセチレンブラック (平均粒径40nm)						
	フェノール樹脂						
導電性接着剤層の厚さ (μ m)							
コア部	金属	ステンレス	ステンレス		アルミ板	アルミメッシュ	
	樹脂				液晶ポリマー		
スキン部	熱導金属				金		
	導電性フィラー	100	100	100		100	膨張黒鉛シート
	樹脂	20	20	5		20	
固有抵抗 (m Ω ·cm)		18	25	408	400	300	300
ガス透過率 (ml/m ² ·day·atm)		2	4	30000	20000	15000	20000
20回繰り返し圧縮後のセパレータ の状態		A	A	B	C(蒸着部分)	B,C(7 μ mメッシュと成形体の界面)	B,C(7 μ mシートと膨張黒鉛シートとの界面)
500時間充電後の電圧値(初期値を100とした場合)		97	94	測定不能	測定不能	測定不能	測定不能
500時間後のセパレータの状態		A	A	B	B	B	B

A: 問題なし。ひび、スキン部の剥離はない。

B: 亀裂が入り、燃料電池セパレータとして使用できなかった。

C: 剥離し、燃料電池セパレータとして使用できなかった。

[0051]

[Effect of the Invention] In this invention, since the conductive core section and the conductive skin section have pasted up by the conductive glue line, even if it is hard to be generated and sheet-metal-izes exfoliation between the conductive core section and the conductive skin section, the reinforcement or gas impermeability do not fall.

[0052] Moreover, the conductive core section and the conductive skin section have pasted up by the conductive glue line, namely, since the conductive core section and the conductive skin section have stuck, the stopped thing of the increment in the resistance as a separator for fuel cells by the increment in contact resistance is made.

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is the sectional view showing an example of the structure of the separator for fuel cells.

[Drawing 2] It is the conceptual diagram showing an example of the manufacture approach of the separator for fuel cells.

[Drawing 3] It is the conceptual diagram showing an example of the manufacture approach of the separator for fuel cells.

[Drawing 4] It is the conceptual diagram showing an example of the manufacture approach of the separator for fuel cells.

[Description of Notations]

- 1 Conductive Core Section
- 2 Conductive Glue Line
- 3 Conductive Skin Section
- 4 Circulation Way of Reactant Gas

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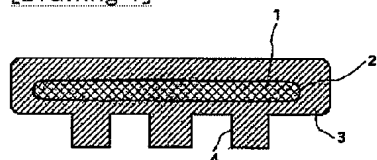
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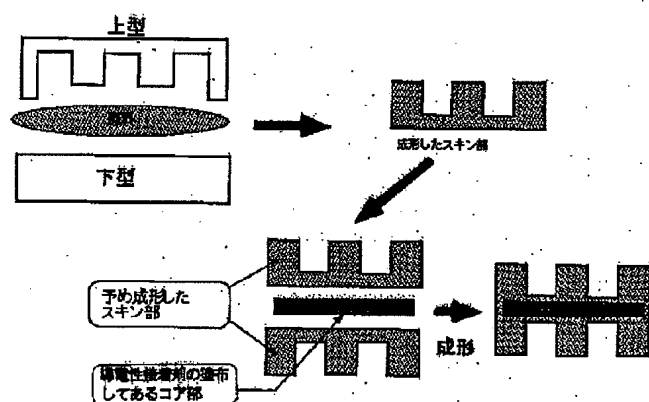
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DRAWINGS

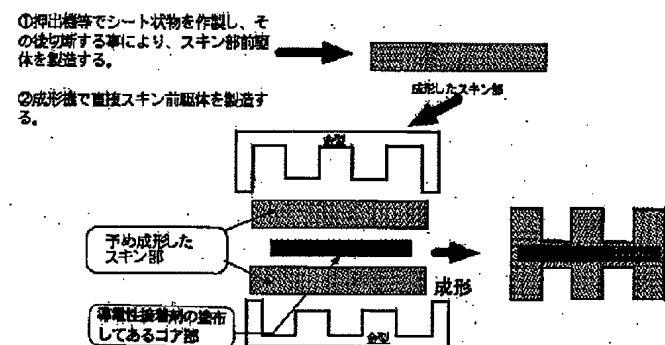
[Drawing 1]



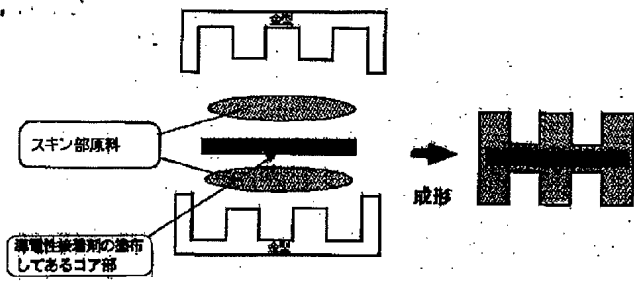
[Drawing 2]



[Drawing 3]



[Drawing 4]



[Translation done.]